Environ Sci Pollut Res (2017) 24:17500–17510
DOI 10.1007/s11356-017-9223-y

RESEARCH ARTICLE

Cancer risk from gaseous carbonyl compounds in indoor environment generated from household coal combustion in Xuanwei, China

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Received: 25 October 2016 / Accepted: 8 May 2017 / Published online: 7 June 2017

Abstract Airborne carbonyls were characterized from emitted indoor coal combustion. Samples were collected in Xuanwei (Yunnan Province), a region in China with a high rate of lung cancer. Eleven of 19 types of samples (58%) demonstrated formaldehyde concentrations higher than the World Health Organization exposure limit (a 30-min average of 100 μg m^{-3}). Different positive significant correlations between glyoxal/methylglyoxal and formaldehyde/acetaldehyde concentrations were observed, suggesting possible different characteristics in emissions between two pairs of carbonyl compounds. A sample in the highest inhalation risk shows 29.2 times higher risk than the lowest sample, suggesting different coal sampling locations could contribute to the variation of inhalation risk. Inhabitants in Xuanwei also tend to spend more time cooking and more days per year indoors than the national average. The calculated cancer risk ranged from 2.2–

Highlights
- Over 50% of samples produce higher formaldehyde concentrations than the exposure limit
- Positive significant correlations show different emission characteristics
- A sample with the highest inhalation risk is 29.2 times higher than the lowest sample
- Over 60% of samples indicate cooking risks at a high level

Responsible editor: Philippe Garrigues

Electronic supplementary material The online version of this article (doi:10.1007/s11356-017-9223-y) contains supplementary material, which is available to authorized users.

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63×10^{-5}, which shows 13 types of samples at high-risk level. Cumulative effect in combination with different carbonyls could have contributed to the additive actual inhalation cancer risk. There is a need to explicitly address the health effects of environmentally relevant doses, considering life-long exposure in indoor dwellings.

Keywords Carbonyl · Indoor air · Coal · Cancer risk

Introduction

Coal is a major energy source. Coal combustion accounts for ~25% energy consumption worldwide (Zhang et al. 2008), and China has become the largest consumer of coal in the world (World energy outlook 2012; Lin and Ouyang 2014). China is also a large energy consumer, and over 75% of its electricity supply comes from coal combustion (Liu et al. 2008). The country is facing severe carbonaceous aerosol pollution and suffering from frequent haze events as a result of deteriorated environmental quality (Tao et al. 2016; Tie and Cao 2009).

Xuanwei County is located in the Yunnan Province of China with a population approximately 1.2 million living in an area of 6257 km^2. The county is one of the major coal-producing regions in Yunnan and renowned for an exceptionally high lung cancer rate (i.e., 2004–2005 lung cancer mortality rate: Xuanwei, 91 per 100,000; national average; 31 per 100,000) (Lin et al. 2015). Past studies linked lung cancer mortality with coal combustion emissions in the area (Barone-Adesi et al. 2012; Kim et al. 2014; Mumford et al. 1987, 1993). The organic extracts of smoky coal samples regularly used in the area were proven to be mutagens, and these samples were shown to be a mouse skin carcinogen and potent initiator of skin tumors (Mumford et al. 1989, 1990). The relevant coal burning studies usually concentrated on characterizing polycyclic aromatic hydrocarbons (PAHs), mineralogical compositions, household fire pit for the emissions, etc. (Chuang et al. 1992; Dai et al. 2008; Mumford et al. 1995; Tian et al. 2008). A past study presented emissions of carbonyl compounds for commonly used cookstoves in China (Zhang and Smith 1999). However, the role of carbonyl compounds in the coal smoke at Xuanwei has been largely overlooked.

Airborne carbonyls (aldehydes and ketones) have been attracting the attention of atmospheric scientists over the past few decades. Carbonyl compounds are identified with natural and anthropogenic sources. The compounds can further be produced via primary and secondary source formation such as incomplete combustion of fossil fuels and biomass, industrial emission, vehicular exhaust, and photochemical oxidation of atmospheric hydrocarbons (Atkinson 2000; Carlier et al. 1986; Grosjean et al. 2002; Kean et al. 2001; Lee et al. 1997; Perry and Gee 1995; Yokelson et al. 1999). The lifetimes of airborne carbonyls are short in the troposphere (De Smedt et al. 2008; Wert et al. 2003), but nevertheless, airborne carbonyls are able to undergo rapid photolysis and generate significant amounts of free radicals and precursors responsible for air pollution (e.g., secondary organic aerosol (SOA) and ozone (O_3) formation) (Carter 1994). Several carbonyl compounds are widely accepted as toxic air contaminants and contain potential carcinogenic and mutagenic properties (CEPA 1993; McLaughlin 1994; NCR 1981; Pal et al. 2008; Seco et al. 2007; WHO 2000).

Formaldehyde is a human carcinogen (group 1) (IARC 2006) and poses nasopharyngeal cancer (IARC 2004). Repeated occupational exposure to formaldehyde in a chemical factory could increase opportunities of having a health implication such as congestion in the cornea, nasal membrane, and pharynx (Zhang 1999). Acetaldehyde is a suspected human carcinogen (Báez et al. 2003; Zhang et al. 1994).

Indoor carbonyl concentrations are a concern as people spend over 80% of their lifetime in an indoor environment (Klepeis et al. 2001). Cooking and heating often involve low-grade solid fuel usage (e.g., coal with impurities such as arsenic and mercury) in underprivileged areas (e.g., Xuanwei) in China (IARC 2010). The combustion processes in household coal stoves usually generate gaseous pollutants (e.g., formaldehyde, CO, CO_2, NO_x, and volatile organic compounds (VOCs)) that are subject to indoor air pollution (Zhang and Smith 1999). The present study deduces possible associations between exposure to indoor coal carbonyl emissions and excess lung cancer risks at Xuanwei. The area has been in agony of abnormal lung cancer mortality rate in many years.

The aims of this study are to (1) characterize gaseous-phase carbonyl compounds from 19 types of coal used in Xuanwei, (2) determine and characterize relationships between carbonyl compounds, and (3) estimate the potential health implications of interior coal emissions for local inhabitants.

Materials and methods

Experimental procedures

Indoor environmental conditions mimic inhabitants under real exposure condition in lung cancer epidemic outbreak at Xuanwei ("pre-1990" kitchen design)

The experimental setup intended to mimic the exposure condition in the timeline when inhabitants first started to suffer from the outbreak (worst case scenario) (Mumford et al. 1987, 1989). Nineteen types of coal were tested for emissions. The samples were labeled 1–19 and were collected from different locations as noticed in Table 1. The samples collected from different coal seams were denoted in brackets (Table 1).

Table 1 Descriptive analysis and relative abundances of carbonyl concentrations

Sampling location	C1 μg/m^3	C2	Acetone	nC3	MEK	iso + nC4	benz	iso-C5	nC5	o-tol	m-tol
1 (Huchang)	305.1 ± 311.3	162.0 ± 115.9	71.1 ± 43.2	27.5 ± 19.5	43.1 ± 26.8	15.0 ± 7.1	88.6 ± 62.5	20.2 ± 11.2	20.2 ± 12.7	16.9 ± 11.4	46.5 ± 32.5
2 (Wenxing)	286.3 ± 210.6	93.7 ± 53.9	41.6 ± 8.3	13.0 ± 5.2	20.8 ± 8.0	6.7 ± 2.4	64.0 ± 48.9	9.3 ± 0.8	9.7 ± 2.7	10.6 ± 5.1	39.4 ± 27.7
3 (Dongshan)	201.9 ± 161.5	130.4 ± 59.5	60.4 ± 23.5	25.4 ± 5.6	43.1 ± 13.0	14.7 ± 3.8	70.9 ± 40.9	27.8 ± 3.8	32.8 ± 8.5	18.1 ± 4.4	40.2 ± 13.1
4 (Tianba)	124.8 ± 67.1	114.6 ± 12.6	81.8 ± 4.0	21.8 ± 2.1	40.3 ± 1.9	12.2 ± 0.7	48.7 ± 4.1	18.2 ± 1.8	17.0 ± 2.0	10.5 ± 3.2	26.7 ± 4.8
5 (Dongshan)	69.9 ± 54.8	50.8 ± 41.8	32.8 ± 17.2	9.0 ± 6.4	18.9 ± 12.5	6.3 ± 3.0	27.7 ± 19.9	8.0 ± 3.2	6.1 ± 1.9	2.9 ± 3.0	10.8 ± 8.3
6 (Jiubao)	423.0 ± 208.5	167.1 ± 62.5	54.7 ± 20.5	25.5 ± 12.2	34.8 ± 16.1	14.8 ± 5.8	72.4 ± 21.2	14.5 ± 6.4	14.7 ± 6.7	12.4 ± 6.9	37.8 ± 22.4
7 (Yefei)	502.6 ± 148.8	195.4 ± 40.0	72.5 ± 26.6	30.9 ± 5.5	39.9 ± 9.7	18.4 ± 5.3	87.1 ± 26.7	20.2 ± 9.2	22.2 ± 12.3	18.5 ± 3.1	46.2 ± 9.1
8 (Reshui)	36.4 ± 18.8	58.2 ± 9.4	23.9 ± 7.5	3.2 ± 0.4	4.7 ± 1.3	2.0 ± 0.5	13.7 ± 5.7	1.8 ± 0.4	B.D.	0.9 ± 0.2	5.2 ± 0.8
9 (Laibin)	269.3 ± 128.6	73.1 ± 35.0	38.9 ± 17.3	11.9 ± 4.7	21.3 ± 11.6	7.3 ± 2.9	39.6 ± 15.0	10.7 ± 3.4	9.3 ± 2.7	7.7 ± 1.2	23.5 ± 8.9
10 (Laibin)	250.1 ± 399.8	154.7 ± 134.9	82.3 ± 37.1	24.8 ± 19.8	47.9 ± 32.7	12.1 ± 8.2	79.4 ± 87.4	21.1 ± 9.0	15.5 ± 8.8	14.3 ± 12.9	40.1 ± 40.5
11 (Laibin)	155.4 ± 117.3	80.2 ± 47.6	59.5 ± 29.8	12.5 ± 7.1	32.5 ± 17.3	8.3 ± 4.5	52.2 ± 39.4	13.2 ± 3.6	10.1 ± 4.8	7.1 ± 3.6	23.6 ± 14.2
12 (Laibin)	122.3 ± 20.8	96.5 ± 31.8	74.2 ± 25.1	17.7 ± 4.6	38.2 ± 8.9	8.9 ± 1.9	41.6 ± 6.8	19.0 ± 5.6	13.7 ± 2.3	7.8 ± 1.4	22.2 ± 4.5
13 (Zhaojiachong)	39.6 ± 55.2	83.8 ± 26.4	92.6 ± 39.8	17.0 ± 6.9	46.4 ± 21.2	12.1 ± 6.5	42.3 ± 17.2	27.2 ± 16.2	17.7 ± 11.0	9.7 ± 2.9	23.9 ± 10.4
14 (Laibin)	12.3 ± 4.8	17.0 ± 5.9	17.6 ± 4.1	2.4 ± 0.7	4.3 ± 0.3	1.6 ± 0.3	5.6 ± 0.3	2.3 ± 0.6	B.D.	B.D.	2.1 ± 0.3
15 (Laibin)	51.0 ± 39.8	122.4 ± 11.5	95.0 ± 15.3	24.4 ± 2.6	54.5 ± 3.0	14.3 ± 2.5	61.9 ± 2.9	31.1 ± 6.0	27.5 ± 7.5	12.5 ± 6.0	33.1 ± 7.7
16 (Laibin)	158.3 ± 87.9	82.1 ± 73.8	67.5 ± 18.3	12.6 ± 11.6	23.9 ± 18.6	7.4 ± 5.8	48.7 ± 40.8	10.6 ± 9.1	9.9 ± 10.9	6.6 ± 7.5	21.0 ± 18.9
17 (Longchang)	57.1 ± 48.9	90.1 ± 34.3	75.8 ± 24.8	12.5 ± 4.5	27.3 ± 10.8	7.5 ± 2.6	38.3 ± 15.4	9.8 ± 2.1	7.1 ± 1.0	3.2 ± 1.8	15.3 ± 6.2
18 (Laibin)	10.4 ± 5.9	79.5 ± 38.8	111.3 ± 38.9	16.5 ± 8.2	39.8 ± 12.1	9.5 ± 4.0	39.5 ± 12.9	24.6 ± 6.5	17.6 ± 4.2	12.6 ± 2.0	26.2 ± 7.2
19 (Zhaojiachong)	19.2 ± 12.2	73.6 ± 31.4	86.2 ± 47.4	14.5 ± 4.2	30.9 ± 10.1	8.5 ± 2.5	38.8 ± 19.3	13.7 ± 4.6	12.8 ± 6.7	7.9 ± 5.9	20.2 ± 9.5

Sampling location	p-tol μg/m^3	C6	2,5-DB	C7	C8	C9	gly	mgly	Total carbonyls
1 (Huchang)	19.4 ± 13.6	23.5 ± 19.1	15.4 ± 14.1	15.0 ± 9.2	11.6 ± 7.5	15.0 ± 8.4	18.3 ± 17.1	12.3 ± 11.3	946.7 ± 636.3
2 (Wenxing)	12.9 ± 7.1	12.4 ± 6.3	7.9 ± 1.3	10.5 ± 6.1	10.0 ± 4.3	16.3 ± 3.1	18.6 ± 6.3	13.2 ± 5.4	697.1 ± 410.0
3 (Dongshan)	18.1 ± 5.5	19.4 ± 5.0	12.8 ± 3.3	17.2 ± 8.0	16.0 ± 3.7	20.6 ± 7.1	7.9 ± 4.5	8.0 ± 5.2	785.7 ± 360.2
4 (Tianba)	10.6 ± 2.2	13.8 ± 3.5	8.4 ± 1.7	12.6 ± 4.9	16.3 ± 4.1	19.1 ± 6.7	5.8 ± 1.4	4.7 ± 1.7	607.9 ± 98.5
5 (Dongshan)	4.0 ± 3.5	7.3 ± 3.4	5.1 ± 1.7	B.D.	7.8 ± 0.7	10.3 ± 1.8	3.0 ± 1.3	2.3 ± 1.2	283.3 ± 184.1
6 (Jiubao)	10.3 ± 4.2	18.1 ± 5.8	6.8 ± 3.8	B.D.	14.1 ± 2.9	15.3 ± 2.8	9.3 ± 5.3	8.6 ± 4.4	954.1 ± 405.9
7 (Yefei)	17.9 ± 2.9	20.4 ± 6.8	11.6 ± 6.6	10.6 ± 3.7	12.7 ± 3.8	13.5 ± 3.1	10.4 ± 6.3	12.5 ± 6.4	1163.6 ± 296.4
8 (Reshui)	2.3 ± 0.3	3.3 ± 0.6	2.2 ± 0.4	B.D.	9.4 ± 2.0	5.5 ± 0.8	0.8 ± 0.4	1.2 ± 0.4	174.8 ± 16.5
9 (Laibin)	9.2 ± 1.5	13.0 ± 5.6	11.5 ± 12.1	8.1 ± 2.9	9.7 ± 0.9	16.2 ± 1.7	8.3 ± 2.9	8.1 ± 3.7	596.6 ± 259.4
10 (Laibin)	16.4 ± 15.7	21.1 ± 18.8	9.1 ± 5.2	13.9 ± 6.7	11.0 ± 4.8	18.6 ± 5.9	10.2 ± 11.0	9.9 ± 11.0	852.7 ± 867.8
11 (Laibin)	10.2 ± 6.3	13.7 ± 6.2	6.7 ± 2.1	14.6 ± 4.5	12.0 ± 4.2	15.9 ± 2.2	5.2 ± 1.0	4.7 ± 2.5	537.5 ± 312.1
12 (Laibin)	9.2 ± 2.2	13.6 ± 3.9	7.3 ± 2.1	16.0 ± 9.6	11.8 ± 4.3	19.3 ± 3.4	8.2 ± 2.3	6.8 ± 2.5	554.3 ± 130.0
13 (Zhaojiachong)	10.1 ± 3.5	11.5 ± 7.1	6.9 ± 4.2	13.3 ± 8.8	9.8 ± 5.7	16.2 ± 8.5	7.1 ± 2.7	5.8 ± 3.2	493.0 ± 201.5
14 (Laibin)	1.1 ± 0.2	2.7 ± 0.1	1.6 ± 0.5	B.D.	7.3 ± 3.2	8.9 ± 1.0	1.2 ± 0.0	1.2 ± 0.6	89.2 ± 17.5
15 (Laibin)	14.6 ± 5.5	17.0 ± 0.8	10.8 ± 2.7	15.0 ± 6.6	12.8 ± 1.5	13.3 ± 3.0	5.0 ± 1.5	5.0 ± 3.1	566.3 ± 28.8
16 (Laibin)	8.9 ± 9.3	12.9 ± 9.4	7.1 ± 5.7	11.6 ± 10.3	11.0 ± 3.1	16.0 ± 0.4	5.8 ± 1.8	6.2 ± 3.2	528.1 ± 327.2
17 (Longchang)	5.5 ± 2.5	8.4 ± 1.5	4.4 ± 2.2	7.7 ± 1.6	10.9 ± 0.6	13.4 ± 0.4	4.3 ± 0.8	3.7 ± 1.9	402.2 ± 152.4
18 (Laibin)	13.9 ± 3.1	14.1 ± 2.8	13.4 ± 2.7	16.0 ± 7.3	15.3 ± 4.8	22.4 ± 12.1	5.3 ± 2.4	3.9 ± 2.0	491.9 ± 125.1
19 (Zhaojiachong)	9.5 ± 5.4	8.7 ± 1.6	8.5 ± 5.4	12.1 ± 4.0	11.0 ± 3.1	15.4 ± 8.4	4.9 ± 1.9	4.0 ± 2.0	400.4 ± 140.7

n = 3 for each type of coal

B.D. below detection limit

Sample 7 and 8 were classified without coal seams as the samples were re-processed from coal lumps collected from the surface in the communities. Sample 10 and 12 were collected from different coal mines. The coal combustion experiment was conducted between November 2012 and January 2013, at a kitchen area in a one-story building in a village called Shangzuosuo (Xuanwei). All doors and windows in the living room were closed during the experiment. The volume of the kitchen was ~42.6 m^3 (5.9 m long × 3.8 m wide × 1.9 m high). The air exchange rate in the kitchen was continuously monitored by measuring the first-order decay of carbon dioxide using a Q-Trak™ indoor air quality monitor (model 8550; TSI, Inc., Shoreview, MN, USA). The air change rate was set as 6.9 h^{-1}.

Preparation of fuels

A laboratory stove (internal diameter of 15 cm; shown in Fig. S1–2 of the Supplementary Material) was used to simulate a fire pit for routine daily burning of coal. The position of the stove during the experiment can be referred to Fig. S3 (Supplementary Material). Larger coal pieces were sieved to retain only samples <5 cm in diameter, to facilitate combustion performance. The stove mass (~7 kg) and coal masses sampled (0.8 ± 0.7 to 1.6 ± 0.1 kg) were monitored throughout the experiment. The coal masses were a random factor and epitomized the usual mass range used for domestic cooking activities.

Burning cycle

Nineteen types of coal samples were used in the combustion tests to collect gaseous samples. Analysis of samples of each type of coal was done in triplicate. The fire was set and kindling (biomass (<2 kg) contained dried sugarcane and corn stock as combustion-supporting agents) was assured. The air was then purged through a stove inlet to provide oxygen for combustion and a chimney was installed over the stove to optimize the chimney effect. When full kindling (the fire's longevity was ensured) had occurred (~5 min after initial ignition), ~2 kg of the coal sample was immediately added to the stove. After 10 min from initial ignition, the remainder of the coal sample was used to fill up the stove. The stove was immediately positioned above the burning coal and remained in place until completion of the experiment. The weight of the stove and coal samples were recorded. All biomass materials were completely removed outdoors, prior to setting the fire. A water pot containing 2 kg of water at room temperature was placed above the stove. Coal lumps could melt and coagulate during combustion, which could extinguish the fire. To simulate cooking in the best possible manner, the fire was stoked and poked at the beginning and at 20-min intervals during the combustion cycle to assure favorable air ventilation through the coal lumps. Additional coal was added to the stove at 20-min intervals, and the weights were recorded throughout the cycle. The water was heated to a boil during the heating process. The complete heating process required 30–60 min depending on the different types of coal (Supplementary Materials: Table S1). The remaining ashes were weighed after each combustion cycle. The combustion cycle was on par with household coal burning activity in Xuanwei (~1 h). The fire was either re-used (for another burning cycle with the same type of coal) or extinguished using a water sprayer. The weight of the coal and water was recorded at 10-min intervals during the experiment.

Sample collection

The air samples were collected in silica cartridges impregnated with acidified 2,4-dinitrophenylhydrazine (DNPH) (Sep-Pak DNPH-silica, 55–105 μm particle size, 125 Å pore size; Waters Corporation, Milford, MA) at a flow rate of 0.7 L min^{-1} using a cartridge sampler. Collection efficiencies were confirmed in the field by sampling carbonyls in two identical cartridges connected in series. Efficiencies were calculated as 100% ($1 - A_b/A_f$), in which A_f and A_b denote the amount of carbonyls collected in the front and back sampling tubes, respectively. No breakthrough was observed in the sampling flow rate and time used. The sampling flow rates were checked in the field at the start and end of each sampling period using a calibrated flow meter (Gilibrator Calibrator; Gilian Instruments, W. Caldwell, NJ). A Teflon filter assembly (Whatman, Clifton, NJ) and ozone scrubber were connected to the front of the DNPH-silica cartridge to remove any particulate matter and prevent possible contamination by ozone (Spaulding et al. 1999). Collocated samples were collected to testify sample collection reproducibility (>95%) in the field. A cartridge was reserved for field blank analysis during each sampling campaign and was handled in the same manner as the other sampling cartridges. The amount of carbonyls detected in the cartridges was corrected for the field blank before conversion to air concentration of carbonyl units. The DNPH-coated cartridges were stored in a refrigerator (<4 °C) prior to analysis.

Carbonyl analysis

A total of 19 carbonyls were quantified, including formaldehyde (C1), acetaldehyde (C2), acetone (acetone), propionaldehyde (nC3), methyl ethyl ketone (MEK), butyraldehyde/isobutyraldehyde (iso + nC4), benzaldehyde (benz), isovaleraldehyde (iso-C5), valeraldehyde (nC5), *o*-tolualdehyde (o-tol), *m*-tolualdehyde (m-tol), *p*-tolualdehyde (p-tol), hexaldehyde (C6), and 2,5-dimethylbenzaldehyde (2,5-DB), heptaldehyde (C7), octaldehyde (C8), nonaldehyde (C9), glyoxal (gly), and methylglyoxal (mgly). Unsaturated carbonyls

such as acrolein and crotonaldehyde were detected but not reported because of their low abundances. Unsaturated carbonyl DNP-hydrazones can react with excess reagent to form adducts, leading to ambiguities in quantification due to chromatographic interferences (e.g., double peaks) and response factor issues (Ho et al. 2011; Schulte-Ladbeck et al. 2001). In-house laboratory experiments demonstrated that collection efficiencies were >93 ± 5% for all target carbonyls under the same flow rate, relative humidity, and temperature. Collection efficiencies for heavy carbonyl compounds (e.g., C6) were recorded to be >96 ± 3%. Each DNPH-coated cartridge was eluted with 2.0 mL acetone-free acetonitrile solution (HPLC/GCMS grade, J&K Scientific Ltd., Ontario, Canada) and transferred to a volumetric flask. Previous studies demonstrated that neither DNPH nor DNPH derivatives remained in the cartridge after elution with 2.0 mL acetone-free acetonitrile solution (Ho et al. 2007). Certified calibration standards for monocarbonyl DNP-hydrazones were purchased from Supelco (Bellefonte, PA) and diluted to a concentration range of 15–3000 μg mL^{-1}. The final volume of each calibration solution was filled up to 2.0 mL with acetonitrile/pyridine (HPLC/GCMS grade, Sigma) at a concentration ratio of 8:2 (*v*/*v*). The calibration curve was linearized, and the correlation of determination (r^2) was >0.999. The calibration standards and cartridge extracts were analyzed by injecting 20 μL of the solution into a high-pressure liquid chromatography (HPLC) system (Series 1200; Agilent Technology, Santa Clara, CA) coupled with a photodiode array detector (DAD). A reversed-phase separation column (4.6 × 250 mm Spheri-5 ODS 5 μm C-18, PerkinElmer, Norwalk, CT) was installed in the HPLC system and operated at room temperature (25 °C). The mobile phase consisted of three solvent mixtures: mixture A, 6:3:1 (*v*/*v*) of water/acetonitrile/tetrahydrofuran; mixture B, 4:6 (*v*/*v*) of water/acetonitrile; and mixture C, acetonitrile. The gradient program was operated first at (80% A)/(20% B) for 1 min, second at a linear gradient of (50% A)/(50% B) for 8 min, third at (100% B) for 10 min, fourth (100% C) for 6 min, and finally at (100% C) for 5 min. The elution rate was 2.0 mL min^{-1}. The absorbance of the 360 and 390 nm wavelengths was applied to identify aliphatic and aromatic carbonyls (e.g., benzaldehyde and tolualdehyde), respectively. Identification and quantification of carbonyl compounds were based on retention time and peak area integration of different carbonyl compounds. The minimum detection limit (MDL) was estimated by analyzing a minimum of seven replicates of standard solution containing analyte at a concentration of 0.015 μg mL^{-1}. The following equation was used to estimate the MDL:

$$\mathrm{MDL} = t_{\left(n-1,1-\infty=99\%\right)} \times s \tag{1}$$

where $t_{(n\,-\,1,\ 1-\infty\,=\,99\%)}$ is Student's *t* distribution value at $n - 1$ degrees of freedom and *s* is the standard deviation of the replicates. The MDLs of the target carbonyls range from 0.002 to 0.010 ng μL^{-1}, which can be translated to 0.169–0.846 μg m^{-3} at a sampling volume of 0.0237 m^3. Measured values, precision, accuracy, and validity were optimized throughout the measurements. Quality assurance was performed to ensure the above attributes were within acceptable limits. A quality control procedure was included to assure a measurement precision of 0.5–3.2% for the measured carbonyls.

Exposure assessment and risk characterization

Residents living in the area are potential receptors of airborne carbonyls. Cancer risk due to exposure to gaseous phase carbonyls was estimated by considering direct inhalation exposure of inhabitants in an indoor environment according to the human health evaluation manual supplemental guidance for inhalation risk assessment (Part F) (U.S.EPA 2011). The cancer risk (CR) of carbonyl compounds can be calculated by the following equations:

$$\mathrm{CR} = \text{slope factor} \times \mathrm{LADD} \tag{2}$$

$$\mathrm{LADD} = \frac{C \times \mathrm{IR} \times \mathrm{AF} \times \mathrm{EF}}{\mathrm{BW} \times \mathrm{AT} \times \mathrm{CV}} \tag{3}$$

where LADD (mg kg^{-1} day^{-1}) is the lifetime average daily dose, *C* (mg m^{-3}) is the pollutant concentration, and IR is the average inhalation rate (m^3 h^{-1}). AF (%) is the absorption fraction (assume 100% absorption) (Cheng et al. 2015). EF is the exposure factor and determined by average duration in indoor (h day^{-1}), average indoor exposure frequency (days), and average life expectancy (years). BW is the average body weight (kg). AT (days) is the average exposure duration for carcinogenic/non-carcinogenic effects. An estimated average exposure duration of 25,550 days (70 years) for carcinogenic effect is applied for the calculation, respectively (Hoddinott and Lee 2000). CV is a conversion factor (from μg to mg). The IR, EF, and BW were calculated based on the information given in the Chinese exposure factors handbook, and the average duration indoors was assumed based on time-activity patterns of cooking status at kitchens in a relevant study in China (Duan 2015; Jiang and Bell 2008). In China, populations in various locations (e.g., inland versus coastal) have different economic conditions, dietary habits, and living styles; thus, location and region is an exposure condition that cannot be ignored. Further information can be referred to Table 2. The slope factor in Eq. (2) is determined by reference dose (RFD, $((\mathrm{mg\ kg^{-1}\ day^{-1}})^{-1})$) for all carbonyl compounds according to the Integrated Risk Information System (U.S.EPA 2015). Only formaldehyde (slope factor = 0.021 $(\mathrm{mg\ kg^{-1}\ day^{-1}})^{-1}$) and acetaldehyde (slope factor = 0.01 $(\mathrm{mg\ kg^{-1}\ day^{-1}})^{-1}$) are considered as carcinogenic substances and therefore provided with slope factors in all measured carbonyl compounds. The parameters used in CR assessment were summarized in Table S2 (Supplementary Material).

Table 2 Information about the Chinese exposure conditions

Exposure factors	Yunnan	National average by provinces
Average inhalation rate (IR) ($m^3\ h^{-1}$)	0.645	0.654
Average duration indoors (h day^{-1})	5.0	2.0
Average indoor exposure frequency (days)	320	221
Average life expectancy (years)	69.54	74.83
Average body weight (BW) (kg)	55.9	60.6
Average exposure duration (AT) (days)	25,550	25,550

The CR value in a range of 1–100 × 10^{-6} is deemed in either acceptable (10^{-6}) or tolerable (10^{-4}) level for regulatory purposes (Hu et al. 2012).

Statistical analysis

All the data were analyzed using SPSS statistic 21.0 (IBM®, New York, NY) or GraphPad Prism software (Version 5 for Windows). Figures were prepared by Origin 6.0 (OriginLab Software Inc.). The exposure assessment and risk characterization ("Exposure assessment and risk characterization" section) were conducted with Microsoft Office Excel 2010 (Microsoft Inc.).

Results and discussion

Characteristics of carbonyl compounds

Concentrations of carbonyl compounds

Table 1 shows the total and individual concentrations of carbonyl compounds in different coal samples. The formaldehyde concentrations are in the range of 10.4 ± 5.9–502.6 ± 148.8 μg m^{-3}. Concentrations for the acetaldehyde range from 17.0 ± 5.9 to 195.4 ± 40.0 μg m^{-3}. According to the World Health Organization (WHO) guideline for the indoor environment, formaldehyde has a 30-min average of 100 μg m^{-3} (WHO 2010). A total of 19 types of samples were analyzed: 11 (58%) demonstrated formaldehyde concentrations higher than the exposure limit. A previous study showed formaldehyde concentrations ranged from 240 to 600 μg m^{-3} in an indoor (30 m^3) cigarette combustion experiment (Grimaldi et al. 1996). Typical indoor formaldehyde and acetaldehyde concentrations could be in the ranges of 10–50 and 5–20 μg m^{-3}, respectively (Sarigiannis et al. 2011). The concentration levels in the present study are akin to those of the combustion experiment. Formaldehyde is the most abundant compound in samples 1–7, 9–12, and 16, accounting for 21–45% of the total measured carbonyls. Acetaldehyde is nevertheless the most abundant compound in sample 8, 13–15, and 17–19 accounting for 16–33% of the total measured carbonyls. The results are consistent with formaldehyde and acetaldehyde as the dominant components in a barbecue charcoal combustion study, and also indicate concentration patterns of these carbonyls could be associated with the inhomogeneous nature of the combustion raw materials (observed high standard deviation of concentrations in some of the sub-samples) (Kabir et al. 2010). A residential coal combustion study in China also demonstrated formaldehyde and acetaldehyde were the most abundant carbonyls in five types of coal (Feng et al. 2010). A study that compared carbonyl emissions using different fuels in a diesel engine showed aldehyde emissions were formed by incomplete oxidation of hydrocarbons. Formaldehyde was the most abundant compound and accounted for over 40%, and the next most abundant, acetaldehyde, ranged from 10 to 30% in composition; these two compounds have similar composition characteristics with those of the present study (He et al. 2009). According to the Agency for Toxic Substances and Disease Registry (ATSDR), formaldehyde is an eye, skin, and respiratory tract irritant. A minimal risk level (MRL) of 8 ppb for respiratory health outcomes was set to be associated with chronic inhalation exposure to formaldehyde (ATSDR 1999). The results show a large proportion of this carbonyl compound was in emissions, indicating that control is possibly required.

Figure 1 shows correlations between the log-transformed concentrations of formaldehyde and acetaldehyde, and glyoxal and methylglyoxal. Both show positive significant correlations ($p < 0.05$); however, only glyoxal and methylglyoxal demonstrate a correlation coefficient >0.80 ($n = 57$). The linear relationship between log-transformed concentrations of individual glyoxal and methylglyoxal suggests the two compounds possibly share similar sources. A lower correlation coefficient >0.40 ($n = 57$) is found between log-transformed concentrations of individual formaldehyde and acetaldehyde; this could potentially be due to the two compounds originating from mixed sources.

A past study showed relative humidity could potentially affect the formaldehyde emissions (Parthasarathy et al. 2011). Water solubility of the carbonyl compounds can vary between species. Species in Fig. 1 show different Henry's law constants (Sander 2015). These factors can possibly contribute to the outcome of the correlations, although these require further investigations. Nonetheless, the limitation in the correlations is that how individual combustion parameters (e.g., flame temperature variations) correlate to the concentration

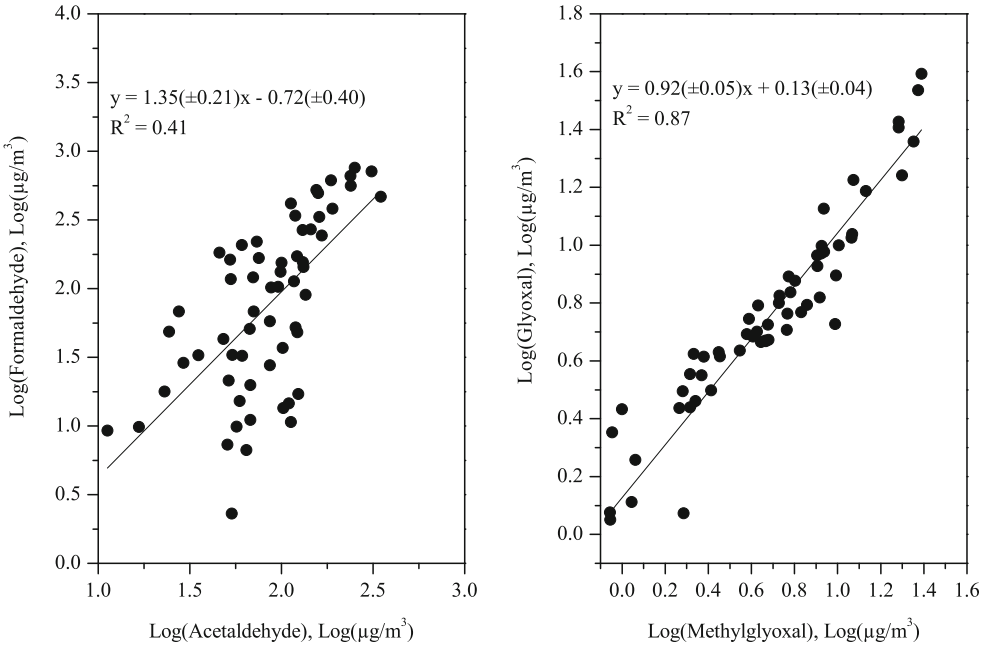

Fig. 1 Relationships between log-transformed concentrations of formaldehyde/acetaldehyde and glyoxal/methylglyoxal. There were 19 types of coal with 3 replicates (n = 57). Coefficients and standard errors were included in the regression equations

variations cannot be determined due to lack of collocated measurements between the two conditions.

The concentration ratios (C1/C2) further show 60% of the samples are formaldehyde emissions dominant over acetaldehyde emissions (>1). The average concentration ratios are in the range of 0.1–3.7. The C1/C2 ratio is a common tool for characterizing pollution sources (Hedberg et al. 2002); the present trend may reflect a variety of contributing factors (e.g., temperature, relative humidity, different coal types, combustion conditions, and sampling procedures), rather than taking account into individual factors alone, could all play different roles altogether in the overall variable outcome. Concentration ratios can be used to compare inter-source similarity. However, the ratios should be applied with caution as the values can vary during the environmental fate of these compounds. The chosen pairs of compounds can be highly reactive and introduce bias to the outcome (Abdullahi et al. 2013).

The strong contribution of original biogenic compounds in the lignite within the early stage of coal formation could ultimately increase the coal rank (Meyer et al. 2014; Püttmann and Schaefer 1990). A previous study suggested the coal combustion process could be divided into three stages: initial stage (moisture evaporation and chemical absorption), combustion stage, and burnout stage—which were classified based on weight and heat changes. Thermogravimetric and differential thermal analysis showed low-rank coals could influence ignition temperatures, whereas high-rank coals influenced the burnout temperature (Moon et al. 2013).

Future studies should be concentrating on characterizing the coal materials (maturity) and different stages of carbonyl emissions such as using proton-transfer-reaction mass-spectrometry (PTR-MS). This technique enables real-time monitoring and is able to instantaneously detect and quantify the emissions, leading to a more thorough understanding about the combustion processes.

Carbonyl emissions from various emission sources

A few studies targeted characterizing carbonyl emissions during emission circumstances. A previous study collected samples in two residential kitchens during a cooking period (used towngas and liquefied petroleum gas). The formaldehyde concentrations were 60.4 and 151.0 μg m^{-3}. Concentrations for the acetaldehyde were 65.9 and 4.5 μg m^{-3}, respectively (Huang et al. 2011). A study investigated concentrations of carbonyl compounds emitted during the combustion of barbecue charcoals and identified formaldehyde and acetaldehyde were the two most abundant carbonyls (Kabir et al. 2010). Formaldehyde and acetaldehyde were the only identified components generated by coal burning in a study that tested different cookstoves in China (Zhang and Smith 1999).

Many studies further characterized background concentrations of carbonyls at different compartments in households. A study measured formaldehyde and acetaldehyde levels in Paris dwellings from potentially different sources in 61 flats with no previous history of complaint for olfactory nuisance or specific symptoms. The result showed average formaldehyde and acetaldehyde concentrations (n = 57) in the kitchen were 21.7 ± 1.9 and 10.1 ± 1.8 μg m^{-3} (Clarisse et al. 2003). A past study targeted domestic levels of formaldehyde in kitchens in 185 homes in Perth, Australia, with a mean concentration of 25.9 μg m^{-3}. The result did not exceed the recommended Australian guideline due to good inter-room

mixing of formaldehyde within homes (Dingle and Franklin 2002). A similar study that measured formaldehyde concentrations in 399 home kitchens in Ankara in Turkey showed the average formaldehyde concentration was 74.9 ± 3.7 μg m^{-3} (Vaizoğlu et al. 2003). A study showed mean indoor concentrations (living room and bedroom) of formaldehyde and acetaldehyde in 16 homes were in the range of 18.1 ± 17.5–46.1 ± 27.3 μg m^{-3} (Marchand et al. 2006). A study measured residences' (71 homes) indoor concentrations of formaldehyde and acetaldehyde in Saskatchewan, Canada. The result suggested in both summer and winter that the formaldehyde and acetaldehyde concentrations were in the range of 10.7 ± 6.4–36.9 ± 18.6 μg m^{-3} (Héroux et al. 2010).

All of the above findings suggest the usual indoor concentrations of formaldehyde and acetaldehyde were below 100 μg m^{-3}, whereas a sample in the present study showed formaldehyde concentrations from the coal emissions could be up to ~5 times, and several samples are at least ~2–3 times higher than the 100 μg m^{-3} level. The present study suggests residential coal combustion at Xuanwei could emit higher formaldehyde concentrations than ordinary indoor levels as mentioned.

Health risk of carbonyl compounds via inhalation exposure

Lifetime excess inhalation cancer risk

Inhalation exposure is typically the primary route of direct exposure to airborne carbonyls. Figure 2 shows the estimated lifetime excess inhalation cancer risk (CR) per million people due to carbonyl exposure in the kitchen in Xuanwei, Yunnan Province. Total carbonyl concentrations in different coal emission samples can be referred to Fig. S4 (Supplementary Material). The non-dietary exposure in this study is defined as human exposure to gaseous carbonyls via household air. Total cancer risk value >10^{-4} is considered to be at high risk in common regulatory programs (Chen and Liao 2006). Under the same carbonyl exposure condition (as in Table 1), the mean estimated excess inhalation cancer risk associated with the exposure is in the range of 22–629 cancer cases per million people (~2.2–63 × 10^{-5}) in the kitchen area at Yunnan. Formaldehyde dominated over acetaldehyde and contributed an average of ~67% of the total risk in all samples. The sample in the highest inhalation risk shows ~29.2 times higher risk than the lowest sample, suggesting different coal sampling locations could contribute to the variation of inhalation risk. Under the same set of PAC emissions, the inhabitants of Yunnan show ~3.6 times higher risk compared to the national average due to different exposure conditions (Table 2) (Duan 2015; Jiang and Bell 2008). A total of 19 types of samples were analyzed: 13 (68%) demonstrated excess inhalation cancer risk higher than the tolerable exposure limit and are classified as high risk. The samples are in the range of ~1.1–6.3 times higher risk than the tolerable threshold level. The carbonyl levels in the kitchen could be an important reference to other living areas in the house especially during winter as all the house windows are usually fastened with limited ventilation; in addition, inhabitants at Xuanwei spend an average of >75% of their time per day indoors (Duan 2015). A cumulative effect in combination with different carbonyls might have contributed to the actual inhalation cancer risk outcome in an

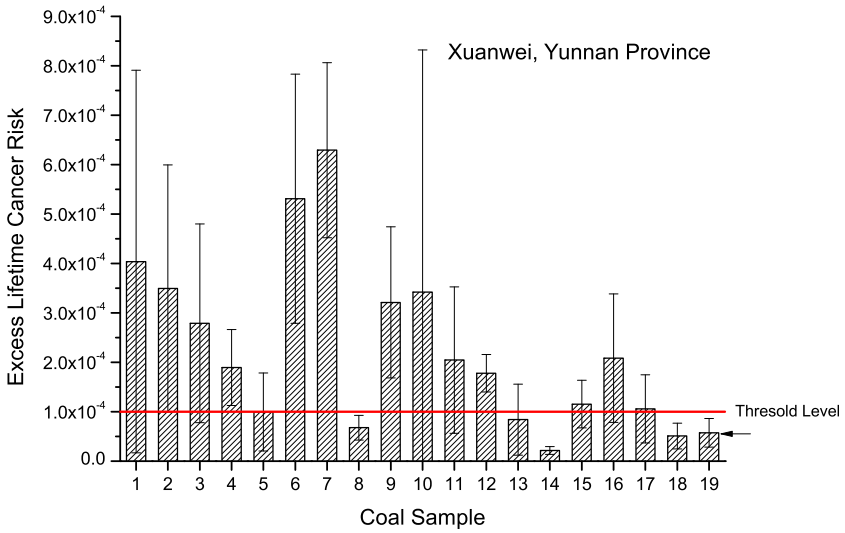

Fig. 2 Column chart for excess cancer risks associated with inhalation exposure of selected carbonyls in coal emissions. *Columns* are shown for inhabitants of the Yunnan exposure scenario. The *arrow* denotes the average value of excess cancer risk of a sample and applies to all samples (samples *1–19*). *Risk error bars* represent standard deviations of the excess cancer risks. The standard deviations were the measured variations of the selected carbonyl concentrations and led to the variations of the risks in analysis. Each type of coal samples was analyzed with three replicates ($n = 3$). *Columns* above the *red line* are classified as within the high-risk level, whereas those below are considered as within the tolerable-risk level

additive manner. Other airborne contaminants (e.g., fine particulate matter and mercury as a coal impurity) during household coal combustion can also pose adverse health effects on humans (IARC 2010; Lui et al. 2017).

The above findings suggest there is a need to revise the current risk assessment in order to explicitly address the health effects of environmentally relevant doses (e.g., absence of carcinogenic risk information except for formaldehyde and acetaldehyde, cancer potency factors in more than binary mixtures), considering the case of life-long exposure in indoor dwellings.

Limitation and uncertainty discussion

Many of the studies on household indoor air pollution have concentrated only on indoor air concentrations without considering personal exposure factors (Clark et al. 2013). The present cancer risk calculation is an attempt to use relevant and accessible information, as the exposure factor is specifically catered for Yunnan Province and only recently launched (Duan 2015). However, the present CR calculation is not without uncertainties. A closer approximation of the actual risks could be produced if a range of weights, inhalation rates, ages, and sex specific for Xuanwei inhabitants were available for the calculations. In addition, insufficient characterization of the sampling households, for example, the number of windows and number of stoves in each household and seasonal variation, could have affected the final cancer risk outcome. Moreover, the limitation of slope factors and reference doses of several targeted carbonyls could have caused a significant under representation of the actual total risk for the analysis. Additional studies should focus on quantifying and harmonizing these uncertainties (e.g., using personal air monitoring devices to collect personal exposure data in Xuanwei households) to improve future cancer risk analyses.

Conclusions

The characteristics of airborne carbonyls emitted during indoor coal combustion in Xuanwei were investigated. Fifty-eight percent of the samples contained formaldehyde concentrations higher than the World Health Organization exposure limit. Positive correlations were identified in a statistical regression analysis, showing possible different emissions characteristics. The lifetime excess cancer risk from inhalation of gaseous carbonyls suggests that 13 types of samples were at high-risk level. Acceptability of the risk depends on scientific data and social, economic, and political factors on the perceived benefits arising from exposure to an agent.

These findings support claims that household coal combustion is associated with human health conditions. The results suggest there is a need to revise the current risk assessment in order to explicitly address the health effects of environmentally relevant doses.

Acknowledgements This study is supported by the project under the Research Grants Council of the Hong Kong Special Administrative Region China (Project No. CUHK 412612). The author would like to thank Xiao-Cui Chen for her assistance in laboratory.

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